Effect of Betaine on self-assembly behavior of some ionic surfactants

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By

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Regards:-

Varsha Rani

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M.Sc. Chemistry honours

CERTIFICATE

Certified that this project report entitled "Effect of Betaine on self-assembly behavior of some ionic surfactants" submitted by Varsha Rani, Registration No: 11603898" student of Physical Sciences, Department of Lovely Professional University, Phagwara, Punjab who carried out the project under my Supervision.

This report has not been submitted to any other university or institute for the award of any degree.

Signature of Supervisor

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ABSTRACT

The main objective of this research work is to gather the information of qualitative understanding of zwitterionic and ionic surfactant by comparing the micellar properties of anionic (SDBS) and cationic (CPC) surfactants in aqueous mixture of Betaine. Conductivity method will be employed to check the micellization behavior of both surfactants in Betaine solution at a temperature range between 298.15K to 308.15K. From these measurements, critical micelle concentration (CMC), degree of dissociation of counter ion (α), standard free energy , standard enthalpy and standard

Entropy of micellization will be calculated. UV-Visible and Fluorescence studies will also be carried out.

KEYWORDS: Zwitterionic surfactant, ionic surfactant, conductivity, Micellization, UV-Visible Spectroscopy

INTRODUCTION

The word surfactant is made up of three words "surface active agents". They mainly lower the surface tension between solid and liquid or two liquids. Surfactants usually play an important role in our daily life with many applications in biology, chemistry and pharmaceutical industries [1]. They are also described as surface active agents. They are the compounds which lower the surface tension between liquid and a solid or two liquids. Any substance that effects the interfacial surface tension can be described as a surfactant but in practical way they can be they can be classified as wetting agents, emulsifiers, agents and dispersants and also plays an important role in drug delivery [2]. In bulk of the solution, these surfactants can assemble into spherical aggregates called as "micelles" and this phenomenon is called Micellization. It is due to diphilic nature of surfactant molecule having spatial arrangement of different system [3]. The concentration at which surfactants first starts to form micelle is known as critical micelle concentration(CMC) which can further be related to thermodynamic behavior of the surfactant.

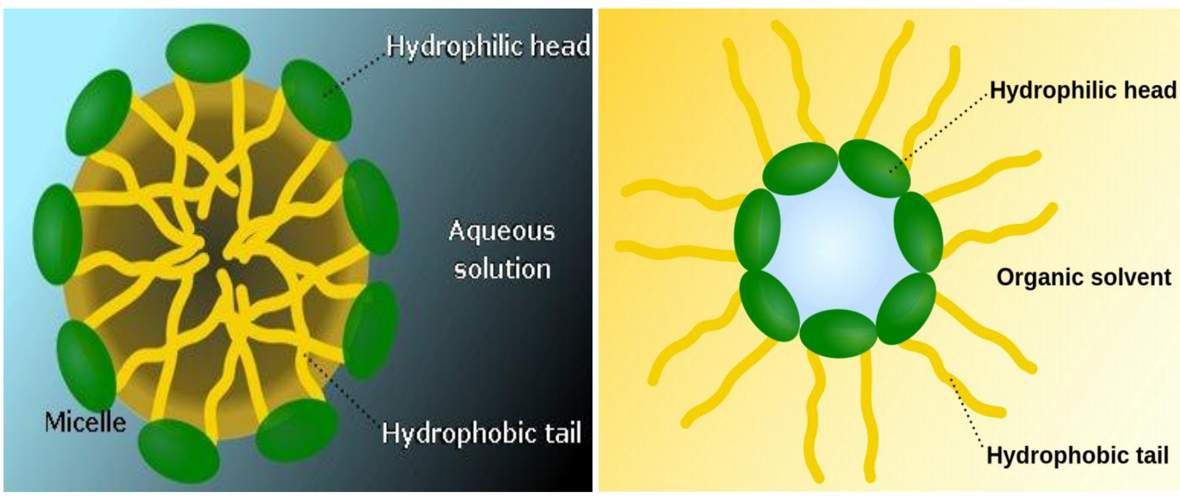

Fig. 1: Representation of surfactant micelles (a) in aqueous solution (b) in organic solvent

Surfactant molecules are formed by two parts with different affinities for different solvents where one affinity is basically for polar and other is for non polar solvent. Whenever micelles are formed inside water, their tales form a core by reducing unfavarouble hydrocarbon- water contacts, and the polar head groups form an outer shell that will maintain favorable contact with water. During the process of micellization, the local orderings of water molecules i.e, hydrophobic hydration is disrupted and the entropy is increased. The shape of molecules mainly depends on the balance in size between hydrophilic head and hydrophobic tail. So, the micellization is entropy driven process. On other side, the head groups are at the core and the tail maintain favorable contact with medium; the aggregate is referred as “reversed type micelle”. It is observed that CMC values for all type of surfactant ranges from 0.5-20 $mmolkg^{-1}$ [4]. In spite of hydrophobic and hydrophilic interactions other factors such as temperature, pressure also effects the behavior of micellization. The physical and chemical properties of diphilic substance have been determined by Traubey’s law. The reactivity of diphilic substance will be triple fold by iny The shape of molecules mainly depends on the balance in size between hydrophilic head and hydrophobic tail introducing $CH_{2\ group}$. As the chain of hydrocarbon increases the large amount of energy is being gained by process of micellization and hence the value of CMC decreases as the length of hydrocarbon will increase.

Classification of surfactants:

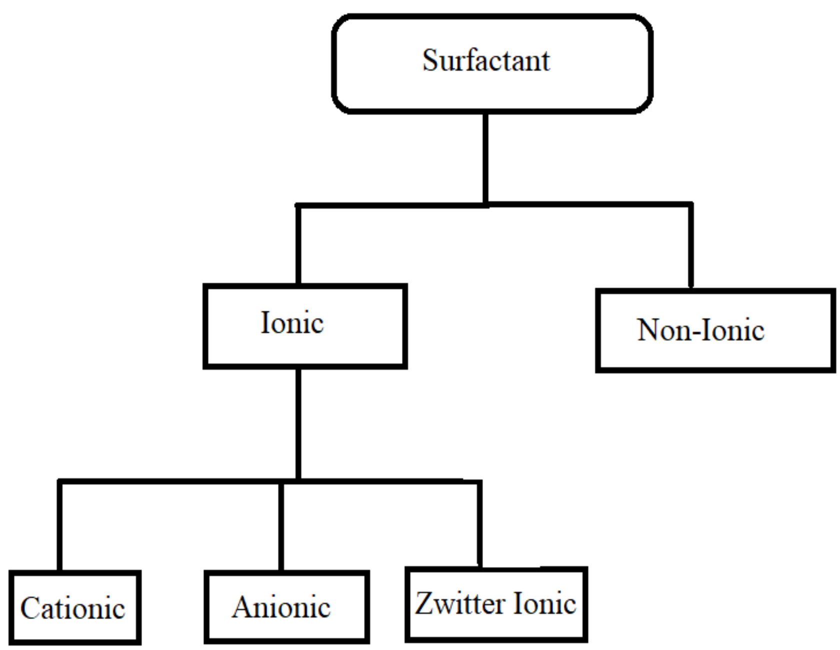

Fig.2: Flowchart showing classification of surfactants

Examples:

Cationic Surfactant:

- Cetyl Pyridinium chloride (CPC)
- CTAB

Anionic Surfactant:

- sodium dodecyl benzene sulfonate SDBS
- sodium dodecyl sulfate SDS

Zwitterionic Surfactant:

- Betaine
- Triton x-100

Non-Ionic Surfactant:

- TWEEN 80
- TWEEN 60

The ionic surfactants selected for the present work are Sodium Dodecyl benzene sulfonic acid (SDBS) an anionic surfactant and a cationic surfactant Cetyl Pyridinium Chloride (CPC). The solution properties of these surfactants have not been critically examined although some reports are available in the literature [5-7].

a) Advantage of selecting sodium dodecyl benzene sulfonate:

This negatively charged surfactant is used in industry as well as household because of their detergency and low cost of manufacture [8-9]. It is also used in tertiary oil recovery process [10].

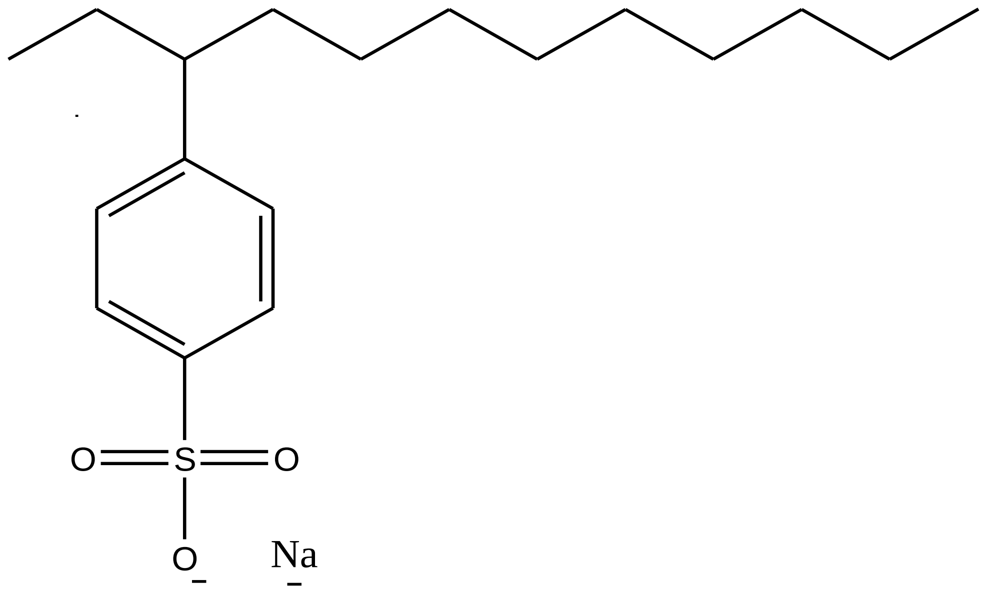

Fig.3: Structure of SDBS(Sodium dodecyl Benzene Sulfonate)

SDBS on dissolving in aqueous medium at low concentration shows specific or cooperative micellization and at higher concentration of surfactant in aqueous medium always cause non specific or non-cooperative micellization. CMC of SDBS is 1.3 mmol kg-1 [11]. It also interacts with cationic and neutral polymers forming solutions at different consistencies.. it also shows antifungal properties and ais also known as good corossion inhibitor [12].

b) Advantages of selecting Cetyl Pyridinium Chloride (CPC)

This cationic surfactant shows antibacterial activity and is even used as softener, lubricant, retarding agent and also cleaning products. It is a commmon component for mouthwash liquids and dental medicines [13]. It has been shown to be effective in preventing dental problems and reducing gingivitis

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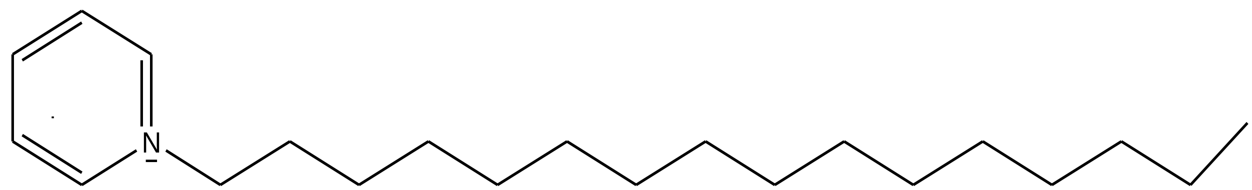

Fig. 4: Structure of CPC (Cetyl Pyridinium Chloride)

CPC is very beneficial in agriculture used in spray solutions because of its low CMC[6]. CMC of CPC is 0.96 mmol kg^{-1} [7-14].

Effect of Betaine on micellization of ionic surfactants:

Micellization behavior of a surfactant differs with respect to temperature and is mainly influenced bu additives[15]. Shape and size of micelles is dependent on the environment surrounding. The micelle and interfacial properties of surfactants are governed by delocate balanace of solvophobic and solvophilic interactions [16].

The results pf studies on effect of betaine on micellar properties of surfactants is due to change in structure of water. If CMC is increasing in presence of additive then it shows decrease in strength of hydrophobic interactions which is considered as water structure breaker. In the same way, decrease in CMC shows increase in hydrophobic interaction and is considered as water structure maker[17].

The study of interfacial and thermodynamic parameters of surfactants in solution can provide extensive information about solue-solute interactions of surfactants in solution. Keeping this view we have important solvent Betaine for our study.

Advantages of selecting Betaine

Betaine is the oxidative product of choline.Biologically betaine serves as organic osmolytes [18]. It is used as a ingredient in toothpaste for reducing the symptoms of dry mouth. It is non-toxic, inexpensive and has a lower viscosity. It mainly binds and stabilises AT base pairs and destabilises GC rich regions. Its addition allows maximal improvement of amplification. It is a methylating agent. It is a topic of interest for its role in osmoregulation. It increases the CMC of ionic surfactants. Betaine protects internal organs, improves vascular risk factors and also enhances performance of the body.

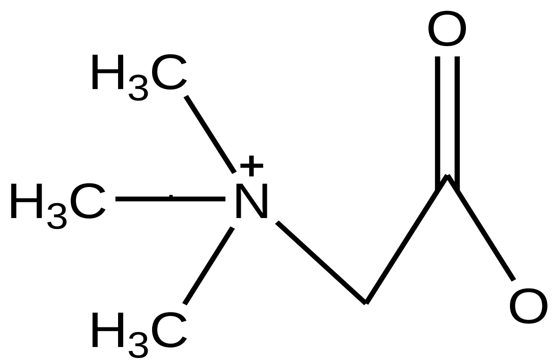

Fig 5 : Structure of Betaine

Literature Survey

The best way to improve the interfacial or surface properties of surfactant is to mix one of the surfactant to another surfactant with which it can interact to produce synergy [19] . It is defined as a condition in which the properties of mixture of surfactants are better than those attainable with individual components of themselves. The study about CMCs of mixed detergents is quite important as it not only helps us to understand about preferential micelle forming tendency of surfactants but even explains about its practical importance [20]. Surface active agents contains impurities of some higher and lower homolouges, and hence the CMC of surfactant would undergo change in presence of impurities and thermodynamics of micellization would also change infact. Rubingh et al propsed the usage of solution theory to calculate the composition of mixed micelle formed by mixing of two surfactants with different head groups in aqueous solutions [20].

Zwitterionic surfactants shows properties that are unique in aqueous solutions, like pH dependent on cmc, stability of foam, lowering of Kraft temperature by salt addition and many more. With respect it is found that the zwitterionic surfactants have more interaction and complez formation with anionic surfactant in aqueous medium. Tsujii et al measured the Kraft points for zwitterionic surfactants of SDS mixed solutions and observed a maximum at some concentrations that was related to intramolecular compound formation in solid phases. Moreover, it was reported that the micellar solution phase of these mixtures of zwitterionic and anionic surfactants displays viscoelastic behavior and extraordinary change in electrical conductivity. Although zwitterionic surfactants have been used as a booster of several anionic surfactants in industrial applications and their mixed properties have been reported, few systematic studies have been carried out up to date. The elucidation of their mechanisms remains to be found [21]. The advancement in the theory of adsorption from solutions of ionic surfactants, and their blends with nonionic ones, gives us detailed analysis and computer modeling of the interfacial properties. The electric double layer development and counter ion adsorption was taken into account [22]. Betaines are used as boosters of foam which provide the property of stabilization of foams against the antifoaming action of oil droplets contained in the commercial shampoos and hair conditioners [23-24]. Moreover, the betaines are known to minimise the irritation action of the surfactant solutions on the eye and skin. Previous studies indicate strong synergistic effects of the mixed solutions of SDS and C_{12} Betaine [23-24]. Synergetic formation of micelles that are rod like in structure has been reported in the mixtures of SDS and cocaamidopropyl betaine, at relatively low surfactant concentration [25]. Dubravka et al showed that the mixed surfactant system shows better results as compared to single surfactant [26-27]. The detergents and the personal care products are mixture of components with various interaction between the constituents that increases the effectiveness of the product, including self-assembly complexes which shows behavior of synergetic. The basic surfactants that are used in personal care products are anionic and amphoteric surfactants to achieve good detergency, quality of cleansing, good foaming properties and easy thickening of salt presence. There are various reports in different literature that gives information about mixed system of surfactants from cationic/cationic, nonionic/nonionic, anionic/nonionic,etc. Behavior of solubilization of different compounds in the mixed micellar solutions was observed that proved to be better than single surfactant micelles. So, detergency is mainly related to micellar stability and addition of surfactants od different charges is the important factor that enhances stability of micelles.

Rio et al studied the temperature effect and chain length of alkyl on different properties of micelle of surfactants using conductometric technique. They also derived even thermodynamic parameters of micellization and found the results according to same [28]. Olofsson and wang studied the micellization behavior of ionic surfactants in aqueous solutions of neutral polymers using micro calometric technique. Sovago et al. concluded that the average orientation of water dipoles pointed toward the bulk. The way their colloidal particles behave in aqueous solutions were investigated within particular temperature range through different methods such as surface tension, light scattering methods and measurements of conductivity [29-33]. Latest studies showed that zwitterionic surfactants can be used in chemical enhanced oil recovery (EOR) because of their great solubility in water, remarkable interfacial properties, high stability of foam, insensitivity to temperature and salinity, and synergistic effect with ionic and anionic surfactants [34-37]. Due to the viscoelastic properties, the surfactants are widely used in many applications, such as personal care products rheology modifiers , heat-transfer fluids, carrier vehicles for drug delivery, micellar catalysts in organic synthesis, templates for preparing mesoporous materials or nanoparticles and in particular, fracturing fluids for improving oil production [37]. Zwitterionic surfactants, whose hydrophilic polar heads carry both a positive and a negative.

The structures of surfactants with hydrophobic groups, betaines can be categorized as alkyl betaines, alkyl amido betaines, α-long chain alkyl betaine, alkoxy betaine, ethoxylated betaine, etc. On basis of acidic radicals, they can be categorized as carboxylates, sulfonates, phosphates etc. As being an important amphoteric surfactant , betaines have outstanding features in various fields such as great surface interface [38-41],good compatibility with other surfactants,low irritation and low biodegradability with certain disinfection and mould proof performance etc.

Proposed Methodology

The methods that we are using are being explained here. Firstly, aqueous stock solution of SDBS and CPC of different molar concentration is made and that is added to the 0.1%, 0.2%, 0.4%, 0.8%, 1.0% of Betaine concentrations, to check the interaction and the micellization behavior of Betaine-SDBS and Betaine-CPC in the aqueous rich mixtures, Conductivity is measured by the conductivity cell. The conductivity cell is calibrated by the 0.1M KCl solution. The cell constant of the conductivity cell is checked and then cell is rinsed with the solution to be used next. Repeating the procedure by altering the temperature from 293.15, 298.15, 303.15, 308.15 (K). When this concentration is done repeat the procedure with the rest of the concentrations taken w/v from 0.1-1%, keeping the concentration of SDBS and CPC constant.

Digital water bath was used to control the temperature. It is a low label water sensor. Digital water bath was supplied by Bombay scientific Pvt. Ltd. The temperature of the digital water bath was maintained within ±0.1℃ over the entire temperature range studied i.e., 293.15K to 308.15K. The temperature of the water bath was also being observed with the help 1/100℃ calibrated thermometer.

Fig 6: Distilled Water Apparatus

CMC Determination by conductivity measurements.

CMC is determined by the conductivity method. A series of stock solution of SDBS and CPC is prepared and added to the Betaine solution by 100 µL capacity. The beaker is gently clamped to the stand and then allowed to attain the temperature which is maintained in the water bath. The conductivity, κ is plotted against the molar concentration of the surfactant and CMC values is being determined as concentration corresponding to the tangents drawn in κ vs. surfactant plots. However, each experiment is carried out at four different temperature 293.15K, 298.15K, 303.15K and 308.15K. Similar procedure is used determination of CMC determination of SDBS and CPC in aqueous rich mixture with the Betaine.

CMC value will be then used to estimate the thermodynamic parameters like standard enthalpy change of micellization ΔH∘m, standard Gibbs free energy of micellization ΔG∘m, standard entropy change of micellization ΔS∘m. The CMC value of SDBS will be determined by the plot of specific conductance (κ) of SDBS in aqueous mixtures of 0.1%, 0.2%, 0.4%, 0.8%, 1% solutions of Betaine at different temperature by conductivity measurements. Different plots of conductance vs. concentration of SDBS are obtained from the readings as per w/v concentrations of Betaine. Before going to calculate the thermodynamic parameters of surfactant-surfactant interaction system we will check for the variation of cmc value of the surfactant at different w/v concentrations of Betaine with the increase in temperature from 293.15K-398.15K.

The CMC data would help us to calculate the standard enthalpy of micellization ΔH∘m of SDBS in aqueous solution of Betaine from the equations.

$$\Delta H_m^o = -RT^2 \frac{d \ln cmc}{dT} \quad (2)$$

Where $\frac{d \ln X_{cmc}}{dT}$ the straight line slope will be obtained by plotting in cmc against T,

The standard entropy of micellization ΔS∘m and standard free energy of micellization ΔG∘m will be calculated by using the following relations

$$\Delta S_m^o = \frac{\Delta H_m^o - \Delta G_m^o}{T} \quad (3)$$

$$\Delta G_m^o = RT \ln cmc \quad (4)$$

Spectroscopic Measurements:

Both the ionic surfactants (SDBS and CPC) selected are UV active and show UV absorbance around λ = (250-260) nm. We will study effect of Betaine on the micellar properties of these ionic surfactant using conductometric technique but in order to get information about nature of surfactant binding with surfactant we will use UV-Vis. Spectroscopy.

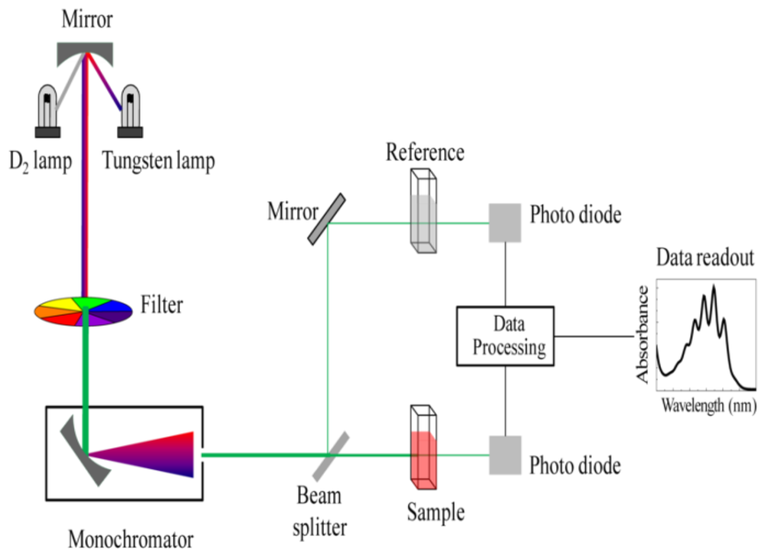

Fig. 7: Schematic representation of working of double beam UV- Vis. Spectrophotometer.

To determine the ground state association constant (K_a) for the interaction of the Betaine and Ionic surfactants we will use Shimadzu UVPC-1800 Spectrophotometer. Benesi-Hildebrand equation will be used to calculate the ground state association constant [42, 43].

$$\frac{1}{\Delta A} = \frac{1}{(\varepsilon_b - \varepsilon_f)} + \frac{1}{(\varepsilon_b - \varepsilon_f) L_T k_a} \frac{1}{M} \quad (1)$$

Where ϵ= extinction coefficient

M= concentration of Betaine

b, f and T= bound states, total ligand and free state.

L_T= the total ligand concentration

ΔA =alteration in absorbance value in a single wavelength.

The value of K_a is estimated from the intercept and the slope of the plot of reciprocal of ΔA versus the reciprocal of concentration of surfactant. We will see spectroscopic behavior of Betaine in Presence of Ionic Surfactants (SDBS and CPC) and Binding efficiency in presence of surfactants .

RESULTS AND DISCUSSIONS:

Table 1: CMC values for SDBS with Betaine at different temperatures.

TEMP	0%	0.1%	0.2%	0.4%	0.8%	1%
273.15	1.2	1.51	1.52	1.62	1.62	1.59
298.15	1.23	1.69	1.54	1.63	1.64	1.64
303.15	1.44	1.81	1.62	1.64	1.72	1.71
308.15	1.46	1.93	1.81	1.72	1.76	1.79

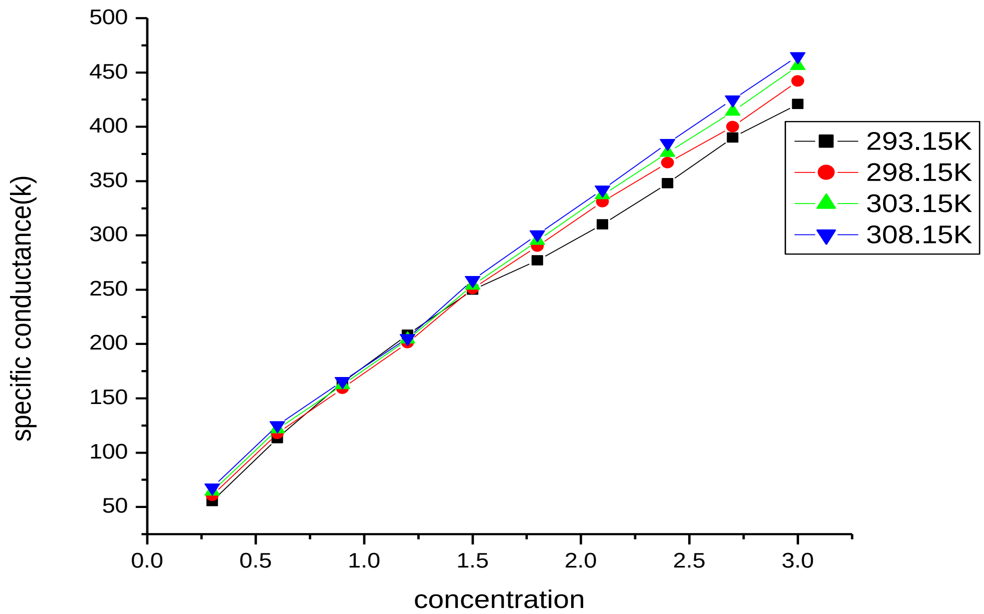

Fig 8: Plot of specific conductance(k) v/s concentration for SDBS and 0% Betaine at different temperatures.

Table 2: CMC values for SDBS with Betaine at different temperatures.

TEMP	0%	0.1%	0.2%	0.4%	0.8%	1%
273.15	0.98	1.01	1.19	1.21	1.12	1.35
298.15	1.00	1.21	1.23	1.29	1.29	1.36
303.15	1.02	1.22	1.26	1.31	1.34	1.39
308.15	1.06	1.24	1.31	1.34	1.41	1.43

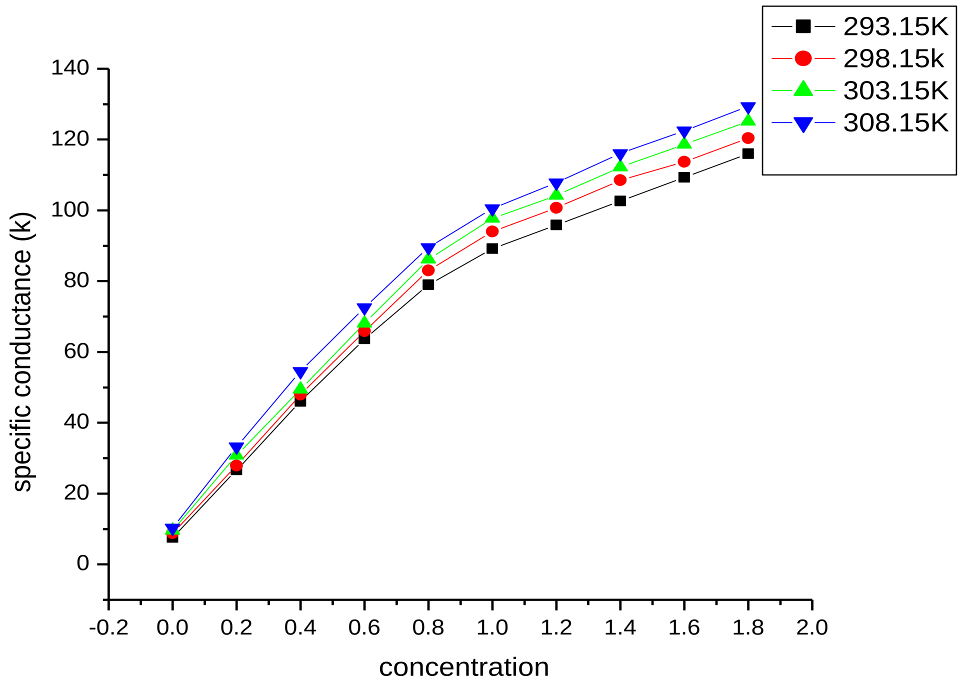

Fig 9: Plot of specific conductance (k) v/s concentration for CPC-0% Betaine concentration at different temperatures.

The present study mainly concluded that CMCs values for SDBS as well as CPC are inceasing with respect to concentration as well as temperature conditions which signify that CMC was directly proportional to degree of disruption of ordered micellar structures. The value of ΔG_m^o will show the presence of electrostatic forces.

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